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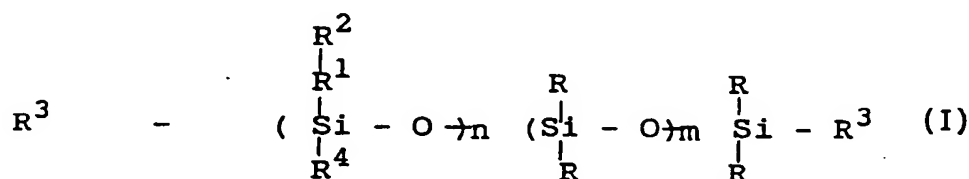
WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification <sup>5</sup> : <b>C08G 77/04, G02B 1/04</b></p>	<p><b>A1</b></p>	<p>(11) International Publication Number: <b>WO 93/21258</b> (43) International Publication Date: 28 October 1993 (28.10.93)</p>
<p>(21) International Application Number: PCT/US93/03497 (22) International Filing Date: 13 April 1993 (13.04.93) (30) Priority data: 07/868,412 14 April 1992 (14.04.92) US (71) Applicant: ALLERGAN, INC. [US/US]; 2525 Dupont Drive, Post Office Box 19534, Irvine, CA 92713-9534 (US). (72) Inventor: YANG, Shih-Liang, Stanley ; 25951 La Cuesta Avenue, Laguna Hills, CA 92653 (US). (74) Agents: VOET, Martin, A. et al.; Allergan, Inc., 2525 Dupont Drive, Post Office Box 19534, Irvine, CA 92713-9534 (US).</p>		<p>(81) Designated States: AU, CA, JP, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>

(54) Title: POLYSILOXANES, METHODS OF MAKING SAME AND HIGH REFRACTIVE INDEX SILICONES MADE FROM SAME



(57) Abstract

A new class of polysiloxanes is disclosed. Such polysiloxanes have formula (I) wherein each R and R<sup>4</sup> is independently selected from alkyl radicals, substituted alkyl radicals, aryl radicals and substituted aryl radicals, each R<sup>1</sup> is independently selected from divalent radicals; each R<sup>2</sup> is independently selected from aryl radicals and substituted aryl radicals; each R<sup>3</sup> is independently selected from monovalent hydrocarbon radicals having a multiple bond and monovalent substituted hydrocarbon radicals having a multiple bond; n is integer in the range of about 6 to about 500; and m is an integer in the range of 0 to about 500. Methods for making such polysiloxanes are also disclosed. In addition, high refractive index, that is having a refractive index of at least about 1.46, silicone compositions useful in the fabrication of lens bodies are disclosed.

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POLYSILOXANES, METHODS OF  
MAKING SAME AND HIGH  
REFRACTIVE INDEX SILICONES  
MADE FROM SAME

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Background of the Invention

The present invention relates to polysiloxanes, methods for producing such polysiloxanes and high refractive index silicone materials made from such polysiloxanes, for example, for use in intraocular lenses. More particularly, the invention relates to polysiloxanes which are structured and are produced to provide silicone materials which have an advantageous combination of properties, including a high refractive index, and are useful in intraocular lenses.

Intraocular lenses (IOLs) have been known for a long time. Such lenses are surgically implanted into the human eye to replace damaged or diseased lenses of the eye.

Whereas IOLs can be made from "hard" polymeric or glass optical materials, soft resilient polymeric materials comprising polysiloxane polymers or copolymers have been increasingly used in the art for this purpose.

IOLs made from silicone polymeric materials are preferably deformable, so that for implantation a smaller incision needs to be surgically cut in the eye than for the implantation of "hard" IOLs. In this respect, the size and mechanical characteristics of the silicone polymeric IOLs play an important role. As it will be well understood by those skilled in the art, for successful implantation the lens must have sufficient structural integrity, elasticity and elongation and small enough size to permit the

folding for insertion through a small incision. After insertion, the lens must, of course, regain its original molded shape and have sufficient structural integrity to retain such shape under normal use conditions.

5 It will be further understood by those skilled in the art that the thinner is the lens, the easier is the surgical insertion procedure. On the other hand, in order to function as an IOL, the lens material must have sufficient optical refractory power. Consequently, the higher is the optical refractive index of the silicone material, the thinner can be the lens to obtain the same optical refractory power.

10 15 Some silicone polymeric materials described in the prior art contain a reinforcer distributed in the polymeric silicone resin. Usually such reinforcement of the silicone polymeric material is necessary for the polymeric material to attain adequate structural strength to be used as a foldable IOL. Examples of reinforced silicone resins suitable for use as soft contact lenses or IOLs are described in United States Patent Nos. 3,996,187; 4,615,702; 3,996,189.

20 25 Travnicek U.S. Patent 3,996,189 discloses that the inclusion of diphenyl siloxane or phenyl-methyl siloxane into a polysiloxane increases the refractive index of the polymer. However, using such phenyl-containing siloxanes as refractive index increasing components results in a polymer which has reduced flexibility or elongation. Thus, although the refractive index is advantageously increased, the elongation of the polymer (and the foldability of an IOL produced from such polymer) is disadvantageously decreased. It would be advantageous to provide a

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siloxane polymer with high refractive index and sufficient elongation to provide a foldable IOL.

Koziol et al U.S. Patent 4,615,702 discloses IOLs made from silicone polymers obtained by polymerization of such monomers as octamethyl-  
5 cyclotetrasiloxane, octaphenylcyclotetrasiloxane, trimethyltriphenylcyclotrisiloxane, and divinyltetramethyldisiloxane. Although such IOLs may have high refractive indexes because of the  
10 relatively high concentrations of phenyl groups, they also disadvantageously have reduced elongation, as do the materials disclosed by Travnicek U.S. Patent 3,996,189, noted above.

Canadian Patent 1,273,144 discloses the inclusion of refractive index modifying groups, such as phenyl groups, into hydride-containing siloxanes by reacting a portion of the hydride groups with carbon-carbon unsaturation bonded to the refractive index modifying group. After this reaction, the  
15 unreacted hydride groups of the modified hydride-containing siloxane are reacted with a compound having at least two carbon-carbon unsaturated bonds to form a cross-linked polysiloxane. This system is somewhat difficult to control and may not be suitable  
20 for mass production of silicone lenses because of potentially large batch-to-batch quality variations. For example, the refractive index modifying groups should be sufficiently numerous and evenly distributed in the hydride-containing siloxane to  
25 provide the desired refractive index without detrimentally affecting the other properties of the final polymer. At the same time, the unreacted hydride groups remaining on the siloxane must be sufficiently numerous and evenly distributed to  
30

provide for the desired cross-linking reaction. These factors can create a reaction control problem which may result in a final polymer not having the desired refractive index and/or not having one or more other desired physical properties. It would be advantageous to provide for increasing the refractive index of a polymer with little or no effect on the cross-linking of the final polymer.

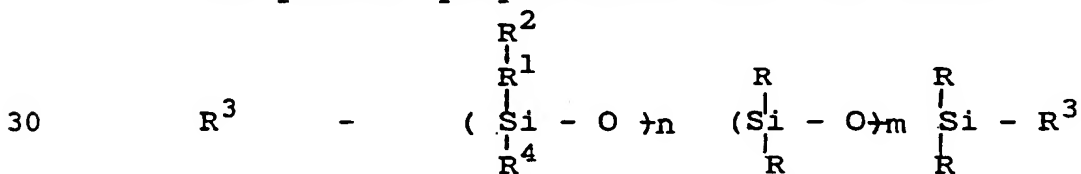
Mbah U.S. Patent 4,882,398 discloses the presence of up to about 40% by weight of a resinous organo-siloxane copolymer consisting essentially of trimethyl siloxy, dimethyl vinyl siloxy and SiO<sub>2</sub> units in a diorgano vinyl siloxy-terminated polydiorgano siloxane containing at least 95 mole% of dimethyl siloxane units and having a viscosity greater than about 12 Pa.s at 25° C decreases the viscosity of the mixture relative to the viscosity of the polydiorgano siloxane. Although this patent does disclose certain aryl and aralkyl groups attached or bonded to a siloxane, there is no teaching or suggestion of any effect on the refractive index of the final polymer as a result of the inclusion of the monovalent hydrocarbon radicals or monovalent halogenated hydrocarbon radicals. Also, the amount of these groups which is included is such as to have little or no effect on the refractive index of the final polymer.

#### Summary of the Invention

New polysiloxanes, methods for making such polysiloxanes, cross-linked copolymers derived from such polysiloxanes, and lenses, for example, IOLs, made from such cross-linked copolymers have been discovered. The present polysiloxanes can be produced in a relatively easy and straight forward

manner. The cross-linked copolymers derived from such polysiloxanes have a very advantageous combinations of properties. In the past, a high content of phenyl groups in a siloxane elastomer, such as a polymethylphenyl siloxane, did increase the refractive index, but also disadvantageously drastically altered the other physical properties, such as elongation and modulus, of the material. The present invention takes advantage of aryl-containing groups to increase refractive index, while reducing or even eliminating the disadvantageous effects that the presence of phenyl groups has heretofore had on the properties of polysiloxane elastomeric compositions. Because these disadvantages are reduced or eliminated, an increased concentration of aryl-containing groups can be included, thus further enhancing the refractive index of the final elastomeric product. For example, cross-linked copolymers produced from the present polysiloxanes can provide optically clear compositions which have high refractive indexes, preferably about 1.46 and above, and are sufficiently flexible so that IOLs can be produced from such compositions which are foldable so as to be inserted into a human eye through a small surgical incision, for example on the order of about 3 mm.

The present polysiloxanes have the formula



wherein each R and R<sup>4</sup> is independently selected from the group consisting of alkyl radicals, substituted alkyl radicals, aryl radicals and substituted aryl

radicals; each  $R^1$  is independently selected from the group consisting of divalent radicals; each  $R^2$  is independently selected from aryl radicals and substituted aryl radicals; each  $R^3$  is independently selected from the group consisting of monovalent hydrocarbon radicals having a multiple bond and substituted monovalent hydrocarbon radicals having a multiple bond;  $n$  is an integer in the range of about 6 to about 500; and  $m$  is an integer in the range of 0 to about 500.

Compositions, for example, elastomeric compositions, in accordance with the present invention comprise a cross-linked copolymer of at least one of the above-noted polysiloxanes and at least one cross-linker component, for example, a polyorgano hydrogen siloxane. Such compositions, which preferably further comprise at least one reinforcer component in an amount effective to increase the strength of the composition, are preferably optically clear and have a refractive index of at least about 1.46. Such compositions may be used to produce lens bodies, for example, optics for IOLs, for use in or on a mammalian eye.

Methods for making polysiloxanes, such as the polysiloxanes described above, are also disclosed. Such methods take advantage of cyclic hydride-containing siloxane monomers, a number of which are commercially available, as starting materials. These cyclic siloxane monomers have well defined compositions so that the concentration of aryl-containing groups substituted for the hydride groups can be easily and very effectively controlled, resulting in polysiloxanes having effectively controlled and distributed aryl-containing



substituents.

Detailed Description of the Invention

The present polysiloxanes have the formula as described above. The amount of aryl-containing substituents in such polysiloxanes is preferably controlled to provide a polysiloxane and/or a cross-linked elastomeric composition derived from such polysiloxane with the desired refractive index, preferably at least about 1.46, more preferably at least about 1.48 and still more preferably at least about 1.50. The aryl-containing substituents preferably are present in such polysiloxanes in an amount of at least about 10 mol%, more preferably at least about 15 mol% and still more preferably at least about 20 mol%, of the total silicon-bound substituents in such polysiloxane. The aryl-containing substituents may be as much as about 40 mol% or about 50 mol% or more of the total silicon-bound substituents in such polysiloxane. In one particularly useful embodiment, substantially all of the aryl-containing substituents are the  $-R_1-R_2$  groups. In this embodiment, each  $R^4$ , and more preferably each R and  $R^4$ , is independently selected from alkyl radicals and substituted alkyl radicals. Still more preferably, each  $R_4$  and R is methyl.

Among the alkyl radicals useful in the present polysiloxanes are those which include 1 to about 10 carbon atoms, preferably 1 to about 4 carbon atoms. Examples include methyl, ethyl, propyl, butyl, octyl and decyl. These alkyl radicals may be substituted with substantially non-interfering substituents which have no substantial detrimental effect on the resulting polysiloxane or on the elastomeric

composition produced from such polysiloxane. Such substituents may include one or more elements, such as oxygen, nitrogen, carbon, hydrogen, halogen, sulfur, phosphorus, and the like and mixtures and combinations thereof.

Each  $R^2$  preferably has 6 to about 10 carbon atoms. The substituents which may be included on the aryl ring include hydrocarbon substituents, such as alkyl groups, as well as other substantially non-interfering substituents, as described herein. In one particularly useful embodiment,  $R^2$  is selected from the group consisting of aryl radicals. More preferably, each  $R^2$  is phenyl.

Preferably, each  $R^1$  radical is independently selected from divalent hydrocarbon radicals and divalent substituted hydrocarbon radicals, more preferably having 1 to about 6 carbon atoms. The substituents which may be included on  $R^1$  are selected from substantially non-interfering substituents, as described herein. In a particularly useful embodiment, each  $R^1$  is independently selected from alkylene radicals, more preferably from a methylene radical or an ethylene radical. In any event, it is important in the present invention that the aryl group or substituted aryl group,  $R^2$ , be separated or spaced apart from the silicon atom to which it is most directly bonded by 1 or more atoms. Without wishing to limit the invention to any particular theory of operation, it is believed that the presence of such a spacer group between the aryl group or substituted aryl group and the silicon atom to which it is most directly bonded provides the enhanced flexibility obtained in the present elastomeric compositions, while, at the same time, having little

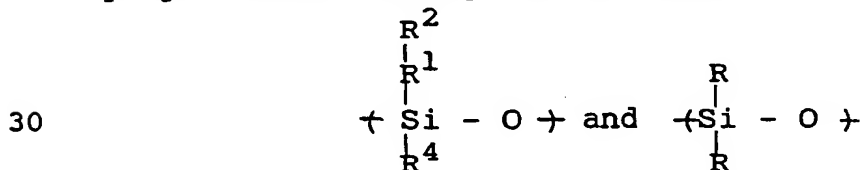
or no adverse effect on the enhanced refractive index achieved by incorporating such aryl-containing groups in the polysiloxane.

Each  $R^3$  preferably has 2 to about 5 carbon atoms, and more preferably includes 1 carbon-carbon double bond. Each  $R^3$  may be substituted with substantially non-interfering substituents, as described herein. More preferably, each  $R^3$  is vinyl.

The present polysiloxanes may be produced by methods which comprise:

(a) contacting a cyclic hydride-containing siloxane monomer with at least one component having the formula  $R^2 - f$  wherein  $R^2$  is as described previously, and  $f$  is a functional monovalent radical capable of reacting with a silicon bonded hydride group of the cyclic hydride-containing siloxane monomer at conditions effective to chemically react the component with at least one of these hydride groups and form a cyclic aryl-containing siloxane monomer containing at least one  $-R^1-R^2$  group wherein  $R^1$  and  $R^2$  are as described previously; and

(b) contacting the cyclic aryl-containing siloxane monomer with at least one siloxane monomer at conditions effective to decyclize and polymerize the cyclic aryl-containing siloxane monomer, polymerize the siloxane monomer and form a polysiloxane having the following units



wherein each  $R$  and  $R^4$  is independently selected as described above.

In another embodiment, methods for making polysiloxanes provide for a step (b) which comprises contacting the cyclic aryl-containing siloxane monomer at conditions effective to decyclize and polymerize the cyclic aryl-containing siloxane monomer and form a polysiloxane having a repeating unit of



wherein each  $\text{R}^4$  is independently selected as described above.

In many instances, at least one precursor of end blocking groups is included in step (b) and is reacted together with the other monomer or monomers present to form the end or terminal groups of the present polysiloxanes. Such precursors, for example, disiloxanes, are such to provide the  $\text{R}^3$  groups to the present polysiloxanes. These precursors preferably include two terminal or end silicon atoms and an  $\text{R}^3$  group bonded to each of the terminal silicon atoms. The precursor or precursors of end blocking groups are preferably included in step (b) in an amount effective to provide the polysiloxane formed with end blocking groups, such as described herein.

In step (a) of the above-noted methods, cyclic hydride-containing siloxane monomers are employed. Such monomers include three or more, preferably 3 to about 6, silicon atoms in a cyclic structure. Such monomers include at least one silicon bonded hydride group, i.e.,  $\text{Si} - \text{H}$ . Preferably, two or more such hydride groups are included in the cyclic hydride-containing monomers. In a particularly useful

embodiment, each of the silicon atoms of such cyclic monomers has at least one, and more preferably only one, hydride group directly bonded thereto. A number of useful cyclic hydride-containing siloxane monomers are commercially available. One especially useful such monomer is tetrahydrotetramethylcyclo-tetrasiloxane.

Step (a) of the present methods may comprise contacting the cyclic hydride-containing siloxane monomer or monomers with at least one component having a formula  $R^2-f$  where  $f$  includes a carbon-carbon multiple, preferably, double, bond at effective hydrosilation conditions.

The contacting at hydrosilation conditions may be catalyzed using, for example, one or more platinum group metal components, many of which are commercially available and conventionally used in vinyl/hydride addition curing of silicone polymers. The amount of platinum group metal, preferably platinum, component employed is effective to promote the desired hydrosilation in step (a). Such amount may be within the range of about 0.01 part per million (ppm) to about 100 ppm (or more) by weight of the total reactants present in step (a), calculated as elemental platinum group metal.

Step (a) may be conducted at hydrosilation conditions effective to provide the desired product. For example, temperatures in the range of about  $10^{\circ}\text{C}$  or lower to about  $60^{\circ}\text{C}$  or higher may be employed. Contacting times in the range of about 10 minutes to about 10 hours or longer have been found to be useful. Since the desired hydrosilation reaction of step (a) is often exothermic, the temperature of the reaction mixture may advantageously be controlled,

e.g., by a cooling medium, to maintain the temperature in the desired range.

Alternately, step (a) can be accomplished using a Grignard-type reaction. In such a reaction, each component  $R^2-f$  is in the form of a so called Grignard reagent, the preparation of which is well known in the art. In this embodiment, step (a) may comprise contacting the cyclic hydride-containing siloxane monomer with a component such a Grignard reagent at effective Grignard reaction conditions to produce the desired cyclic aryl-containing siloxane monomer. Such Grignard reaction conditions include, for example, reaction temperatures in the range of about  $-60^{\circ}\text{C}$  or lower to about  $0^{\circ}\text{C}$  or higher; and reaction times in the range of about 10 minutes to about 10 hours or longer.

The cyclic aryl-containing siloxane monomers alone or with other siloxane monomers, and preferably with precursors of end blocking groups, are reacted in the presence of a suitable catalyst to achieve decyclization and polymerization to the desired degree. The reactions can be conducted by using one or more of a variety of catalysts. Many such catalysts are well known in the art of cyclic siloxane polymerization. Examples of such catalysts include potassium hydroxide, tetramethyl ammonium hydroxide, derivatives thereof and mixtures thereof.

The amount of catalyst used in and the conditions at which step (b) occurs may be similar to those parameters which are conventionally employed in decyclizing and polymerizing other cyclic siloxane monomers. For example, the amount of catalyst employed may be in the range of about 0.01% to about 1% by weight of the total reactants.

Temperatures in the range of about 20° C to about 150° C and reaction times in the range of about 0.5 hours to about 6 hours or more may be employed.

- 5        The degree of polymerization in step (b) is preferably monitored by monitoring the viscosity of the reaction mixture.

After the desired level or degree of polymerization is achieved, the catalyst is  
10        inactivated, neutralized, or removed, and the reaction product may be filtered.

After filtration, volatile materials are removed from the polysiloxane, for example, by repeated vacuum stripping.

- 15        The present elastomeric compositions comprise a cross-linked copolymer of (1) a polysiloxane having a formula as noted above and (2) a cross-linker component. Such elastomeric compositions preferably further comprise a  
20        reinforcer component in an amount effective to increase the strength of the composition. Preferably the present elastomeric compositions are optically clear and have a refractive index of at least about 1.46, more preferably at least  
25        about 1.48 and still more preferably at least about 1.50. Additionally, the present elastomeric compositions preferably have a combination of other properties, such as substantial flexibility, elongation and tensile  
30        strength, to provide optics for IOLs which are foldable and insertable through surgical incisions on the order of about 3 mm.

Particularly useful cross-linker components are selected from polyorganohydrosiloxanes and

mixtures thereof. Many such polyorganohydrosiloxanes are commercially available and/or well known in the art for providing vinyl/hydride addition cure silicone polymers. In order to obtain enhanced compatibility between the polysiloxane and the cross-linker component and and/or enhanced refractive index of the elastomeric material, it is preferred that the cross-linker component have a refractive index which is substantially the same, for example, within about 0.05 and preferably within about 0.02, as the refractive index of the polysiloxane. Examples of particularly useful cross-linker components include copolymers of methylhydrosiloxane and phenylmethylsiloxane, copolymers of methylhydrosiloxane and diphenylsiloxane, and mixtures thereof.

The elastomeric compositions of the present invention preferably contain at least one reinforcer component dispersed in the elastomer composition.

In accordance with one embodiment of the invention, the reinforcer component is preferably used in a ratio of about 1 to about 45 parts by weight of the reinforcer component to 100 parts of the total elastomeric composition. Silica, preferably fume silica, and organic resins are very useful as the reinforcer component. Fume silica itself is commercially available.

Processes for trimethylsilylating the surface of fume silica for the purpose of rendering the silica surface hydrophobic and more compatible with polysiloxane polymers are also known and



within the skill of the art. A number of organic resins are known to be useful for reinforcing articles which include silicone elastomers. Of course, the reinforcer component used in the present elastomeric compositions employed in optical applications should be optically clear or at least have no significant detrimental effect on the optical clarity of the elastomeric composition. The refractive index of the reinforcer component is preferably at least about equal to or greater than the refractive index of the silicone elastomer in the elastomeric composition.

The fume silica reinforcer useful in the present compositions preferably has a surface area of about 100 to about 450 meters<sup>2</sup>/gram.

In the preparation of the present elastomeric compositions, the polysiloxane is preferably intimately mixed with the reinforcer component. The intimate mixing is preferably aided by treating the mixture on a roll mill or like device. After intimate mixing, additional volatiles may be removed from the mixture by heat and vacuum.

This intimate mixture of polysiloxane and reinforcer component is hereinafter referred to as the "base". For the purpose of making materials suitable for use in IOLs, the base may be dispersed in a suitable inert solvent, such as trichlorotrifluoroethane (FREON), and the dispersion filtered to remove any solid impurities. Thereafter, the solvent is removed by gentle heat and vacuum.

The base preferably has the inherent characteristic of providing, after suitable curing by cross-linking, an elastomeric composition having physical properties which are highly advantageous for inclusion in a foldable IOL. Thus, after the curing or cross-linking step, the properties of the resulting cross-linked elastomeric composition preferably include in accordance with the present invention the following:

- an optical refractive index which is at least about 1.46, more preferably at least about 1.48 and still more preferably at least about 1.50;
- a Shore A durometer hardness value of at least about 25;
- a tensile strength of at least about 400 psi;
- a tear strength of at least about 20 pounds per lineal inch (pli); and
- an elongation of at about 100%, preferably at least about 200%.

The above listed properties can be measured in accordance with state-of-the-art technology and instruments in accordance with the respective requirements of standard ASTM test methods. More particularly, the durometer test is performed as ASTM D2240, the tensile and elongation tests as ASTM D412 and the tear strength test as ASTM D624 Die B.

In one embodiment, the durometer hardness is about 38 to about 40, the tensile strength is in the range of about 700 to about 750 psi, and the tear strength is about 40 pli. In this regard it is noted that cross-linking tends to slightly increase the optical refractive index as compared

to the uncured base.

Preparation of the uncured base for cross-linking is preferably accomplished as follows. The base is divided into two aliquots which  
5 preferably are of equal weight. The aliquots are termed "Part A" and "Part B" or first and second aliquot parts. Cross-linking may be accomplished by utilizing a platinum group metal catalyzed reaction of the terminal silicon bonded multiple  
10 bonds (vinyl groups) of the base, and silicon bonded hydrogens or hydride groups of the cross-linking agent. The silicon bonded multiple bonds (vinyl groups) are present in both the first and second aliquots of the base.

15 Silicon bonded hydrogens or hydride groups are added in the practice of the present invention to the second aliquot (Part B) in the form of one or more suitable cross-linking agents, such as a polyorganohydrogen siloxane. The cross-linking  
20 agents per se are known in the art, and may be made in accordance with the teachings of United States Patent No. 3,436,366, which is incorporated in its entirety herein by reference.

The platinum group metal, preferably  
25 platinum, catalyst can be selected from such catalysts which are conventional and well known in the art. Suitable catalysts include organo platinum group metal, preferably platinum, compounds, for example, in accordance with United  
30 States Patent Nos. 2,823,218 and 3,159,601, each of which is incorporated in its entirety herein by reference. The catalyst may be added to the first aliquot (Part A).

After mixing of the aliquots (Part A and Part B), the cross-linking preferably should not proceed too rapidly at room temperature, thereby allowing at least about 2, more preferably at least about 4 or about 6, hours for work time with the mixed aliquots. For this reason, a suitable cross-linking inhibitor, such as 1, 2, 3, 4 tetramethyl- 1,2,3,4-tetravinyl cyclotetrasiloxane, is preferably added to the second aliquot (Part B).

The platinum group metal catalyst is present in the first aliquot in an amount in the range of about 1 ppm to about 50 ppm by weight. The cross-linker is preferably included in the second aliquot in an amount in the range of about 0.5 or about 1 to about 6 or about 10 parts per hundred by weight. The cross-linking inhibitor is preferably added to the second aliquot in an amount in the range of about 0.01 to about 0.2 parts per hundred by weight.

An ultraviolet light absorbing material, preferably a polymerizable ultraviolet light absorbing material, may be mixed into the second aliquot.

The ultraviolet light absorbing material, for example, selected from vinyl functional 2-hydroxybenzophenones and vinyl functional benzotrizoles, is preferably covalently linked to the silicone elastomer of the elastomeric composition during the cross-linking step. The ultraviolet light absorbing material is preferably added to the second aliquot in an amount in the range of about 0.1% to about 1% or about 5% by weight. The curing or cross-linking occurs at

conditions effective to provide the desired elastomeric composition. Curing temperatures may vary, for example, from about 20° C to about 200° C, and curing times may range, for example, from  
5 about 1 minute to about 5 hours or about 10 hours or more.

Formation of IOL bodies or optics from the elastomeric compositions of the present invention may be accomplished by liquid injection molding or  
10 by cast or compression molding or other types of molding of the intimately mixed first and second aliquots. These processes are well known in the art.

The following non-limiting examples  
15 illustrate certain aspects of the present invention.

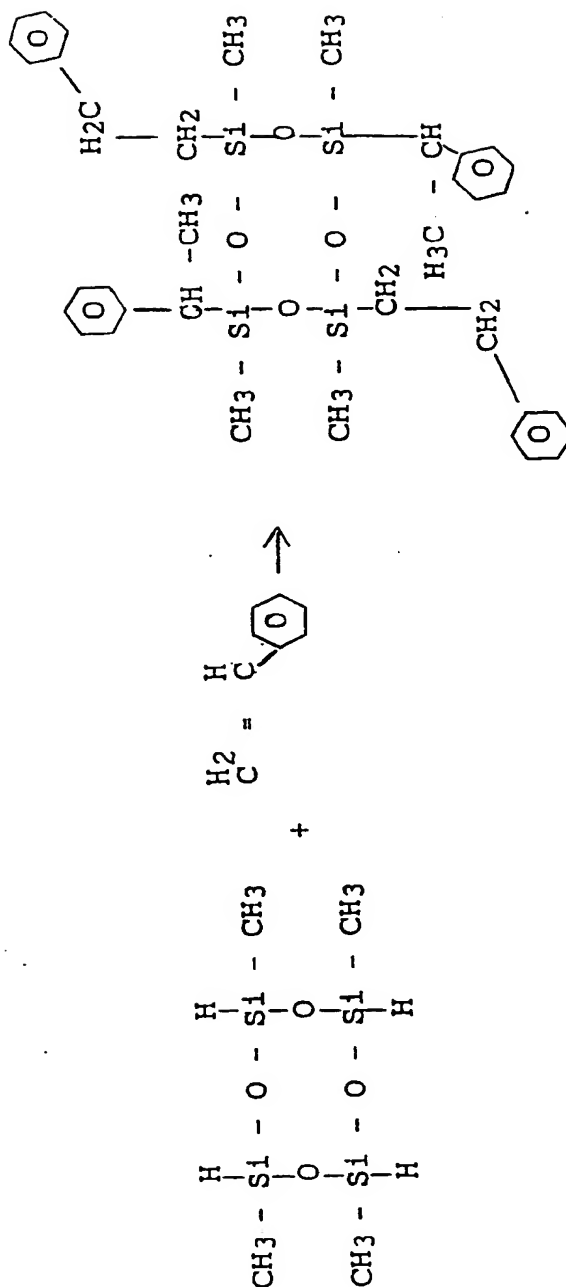
#### EXAMPLE 1

24 g of tetramethylhydrocyclotetrasiloxane (from Petrarch System, Inc.) is added dropwise  
20 into a 25 g toluene solution containing 25 g styrene (Aldrich Chemical Company) and 1 ml platinum complex solution (Petrarch System, Inc.). The reaction is exothermal and is controlled at 40° C using an ice bath. After complete addition  
25 of the tetramethylhydrocyclo- tetrasiloxane in 30 minutes, the reaction temperature is slowly raised from 40° to 75° C in one hour and is maintained at 75° C for another hour. The toluene solvent and unreacted styrene is removed by vacuum  
30 distillation. After the volatiles are removed, the product flask is found to contain 34 g of tetramethylstyrylcyclotetrasiloxane.

"H-NMR analysis indicates that alpha and beta substitutions have a ratio of about 1 to 1.6 and  
35 are as shown in the chemical structure illustrated

below. The tetramethylstyrylcyclotetrasiloxane monomer has a refractive index of 1.53.

The above described reaction is illustrated as follows:



EXAMPLE 2

In a 2 liter reactor, tetramethylstyrylcyclo-  
tetrasiloxane (1088 g) and 1, 2 divinyltetramethyl-  
disiloxane (6 g) are mixed and heated with agitation  
under a nitrogen gas blanket to 100° C. When the  
5 temperature reaches 100° C, 0.18 per cent (by weight)  
N-catalyst (tetramethyl ammonia hydroxide) is added.  
Stirring and heating are continued and the viscosity  
of samples taken from the reaction mixture is  
monitored. If after 45 minutes there is no change in  
10 viscosity, an additional 0.09 percent N-catalyst is  
added. After heating and stirring for another 3  
hours, the catalyst is destroyed by heating the  
mixture to 150° C. The viscosity of the cooled  
reaction mixture should be between 2000 and 2800 cp.  
15 The refractive index should be between 1.52 and 1.54.

The polymer is stripped three times on a wipe  
film evaporator. The viscosity of the stripped  
polymer should be between 4100 and 5300 cp, and the  
refractive index should be between 1.53 and 1.54.  
20 This polymer is a vinyl terminated methyl-  
styrylpolysiloxane.

EXAMPLE 3

The stripped polymer from Example 2 is passed  
through a 325 mesh steel wire screen under pressure.  
25 The batch is divided into two equal parts, Part A and  
Part B. 12 parts per million by weight of the  
organoplatinum catalyst identified in Example 1 is  
mixed into Part A. Small samples from Part B are  
mixed with various concentrations of a cross-linker,  
30 a liquid organohydrogen polysiloxane having a  
refractive index of 1.50 and sold by Petrarch Systems  
under the trademark PS 12905. The cross-linker level



is optimized so as to obtain a Shore durometer hardness of approximately 25 or higher (ASTM D2240) in the cross-linked product. Thereafter, increasing amounts of an inhibitor (1,2,3,4 tetramethyl-1,2,3,4-tetravinyl cyclotetrasiloxane) are added to Part B and mixed samples of Parts A and B are tested to obtain a working time of about 6 hours or longer at room temperature. Depending on the above-noted sample test results, the cross-linker is added to Part B to provide 1-6 parts per hundred by weight, and the inhibitor is added to Part B to provide 0.01 to 0.2 parts per hundred by weight.

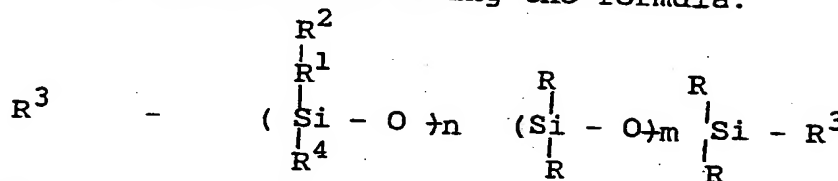
An elastomer prepared by curing equal amounts of Parts A and B (with 6 parts by weight of cross-linker at 100°C for 30 minutes) has a refractive index of about 1.54 and sufficient tensile strength, elongation and tear strength to be useful for making foldable IOLs.

The physical properties of the cured elastomer can be further improved by adding silica gel or resin reinforcing agents into the formulation. Such reinforced elastomeric compositions are very useful for making foldable IOLs.

While this invention has been described with respect to various specific examples and embodiments, it is to be understood that the invention is not limited thereto and that it can be variously practiced within the scope of the following claims.

WHAT IS CLAIMED IS:

1. A polysiloxane having the formula:



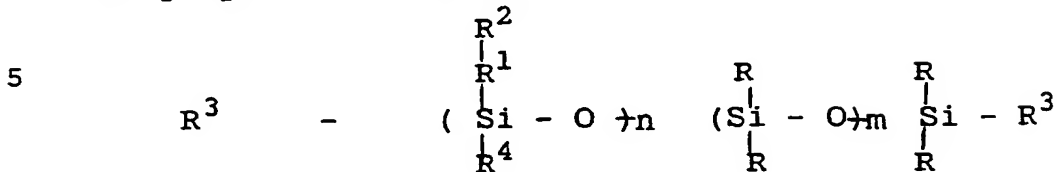
wherein each R and R<sup>4</sup> is independently selected from the group consisting of alkyl radicals, substituted alkyl radicals, aryl radicals and substituted aryl radicals; each R<sup>1</sup> is independently selected from the group consisting of divalent radicals; each R<sup>2</sup> is independently selected from aryl radicals and substituted aryl radicals; each R<sup>3</sup> is independently selected from the group consisting of monovalent hydrocarbon radicals having a multiple bond and substituted monovalent hydrocarbon radicals having a multiple bond; n is an integer in the range of about 6 to about 500; and m is an integer in the range of 0 to about 500.

2. The polysiloxane of claim 1 wherein each R<sup>4</sup> is selected from the group consisting of alkyl radicals and substituted alkyl radicals; each R<sup>2</sup> has 6 to about 10 carbon atoms; and each R<sup>3</sup> has 2 to about 5 carbon atoms.

3. The polysiloxane of claim 1 wherein each R and R<sup>4</sup> is methyl, each R<sup>1</sup> has 1 to about 6 carbon atoms; each R<sup>2</sup> is phenyl and each R<sup>3</sup> is vinyl.

4. The polysiloxane of claim 3 wherein each R<sup>1</sup> is a methylene radical or an ethylene radical.

5. A composition comprising:  
a cross-linked copolymer of (1) at least  
one polysiloxane having the formula



and (2) at least one cross-linker component, wherein  
each R and R<sup>4</sup> is independently selected from the  
group consisting of alkyl radicals, substituted alkyl  
radicals, aryl radicals and substituted aryl  
radicals; each R<sup>1</sup> is independently selected from  
divalent radicals; each R<sup>2</sup> is independently selected  
from the group consisting of aryl radicals and  
substituted aryl radicals; each R<sup>3</sup> is independently  
selected from the group consisting of monovalent  
hydrocarbon radicals having a multiple bond and  
substituted hydrocarbon radicals having a multiple  
bond; n is an integer in the range of about 6 to  
about 500; and m is an integer in the range of 0 to  
about 500.

6. The composition of claim 5 which is  
optically clear and has a refractive index of at  
least about 1.46.

7. The composition of claim 5 which further  
comprises at least one reinforcer component in said  
copolymer in an amount effective to increase the  
strength of said composition.

8. The composition of claim 5 wherein said  
cross-linker component is a poly organo  
hydrosiloxane.

9. The composition of claim 5 wherein each R has 1 to about 3 carbon atoms; each R<sup>2</sup> has 6 to about 10 carbon atoms; each R<sup>3</sup> has 2 to about 5 carbon atoms; and each R<sup>4</sup> is independently selected from the group consisting of alkyl radicals and substituted alkyl radicals.

10. The composition of claim 5 wherein each R and R<sup>4</sup> is methyl, each R<sup>1</sup> has 1 to about 6 carbon atoms, each R<sup>2</sup> is phenyl and each R<sup>3</sup> is vinyl.

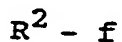
11. The composition of claim 8 wherein said poly organo hydrosiloxane is selected from the group consisting of copolymers of methylhydrosiloxanes and phenylmethylsiloxane, copolymers of methylhydrosiloxane and diphenylsiloxane, and mixtures thereof.

12. A lens body for use in or on a mammalian eye comprising the composition of claim 5.

13. A lens body for use in or on a mammalian eye comprising the composition of claim 7.

14. A method for making a polysiloxane comprising:

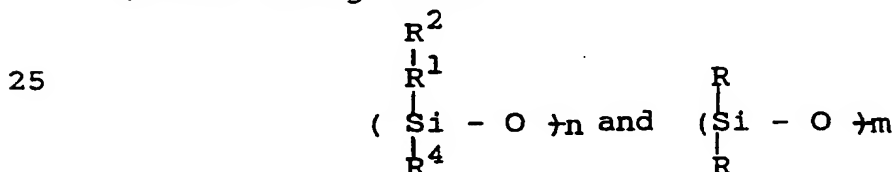
(a) contacting a cyclic hydride-containing siloxane monomer with at least one component having the formula



wherein R<sup>2</sup> is selected from the group consisting of aryl radicals and substituted aryl radicals, and f is a functional monovalent radical capable of reacting with a hydride group of said cyclic hydride-

containing siloxane monomer at conditions effective to chemically react said component with at least one of said hydride groups of said cyclic hydride-containing siloxane monomer and form a cyclic aryl-containing siloxane monomer including at least one - R<sup>1</sup> - R<sup>2</sup> group wherein R<sup>1</sup> is selected from the group consisting of divalent radicals; and

(b) contacting said cyclic aryl-containing siloxane monomer with at least one siloxane monomer at conditions effective to decyclize and polymerize said cyclic aryl-containing siloxane monomer, react said siloxane monomer and form a polysiloxane having the following units



wherein each R and R<sup>4</sup> is independently selected from the group consisting of alkyl radicals, substituted alkyl radicals, aryl radicals and substituted alkyl radicals, n is an integer in the range of 6 to about 500 and m is an integer in the range of 1 to about 500.

15. The method of claim 14 wherein each R has 1 to about 3 carbon atoms; each R<sup>2</sup> has 6 to about 10 carbon atoms; and each R<sup>4</sup> is independently selected from the group consisting of alkyl radicals and substituted alkyl radicals.

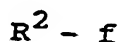
16. The method of claim 14 wherein each R and R<sup>4</sup> is methyl, each R<sup>1</sup> has 1 to about 6 carbon atoms, and each R<sup>2</sup> is phenyl.

17. The method of claim 14 wherein at least one end blocking group precursor is included in step (b) in an amount effective to provide said polysiloxane with end blocking groups.

18. The method of claim 14 wherein said cyclic hydride-containing siloxane monomer has a plurality of hydride groups, and step (a) is effective to chemically react said component with all of said hydride groups of said cyclic hydride-containing siloxane monomer.

19. A method for making a polysiloxane comprising:

(a) contacting a cyclic hydride-containing siloxane monomer with at least one component having the formula



wherein  $R^2$  is selected from the group consisting of aryl radicals and substituted aryl radicals and  $f$  is a functional monovalent radical capable of reacting with a hydride group of said cyclic hydride-containing siloxane monomer at conditions effective to chemically react said component with at least one of said hydride groups of said cyclic hydride-containing siloxane monomer and form a cyclic aryl-containing siloxane monomer including at least one  $R^1 - R^2$  group wherein  $R^1$  is selected from the group consisting of divalent radicals,; and

(b) contacting said cyclic aryl-containing siloxane monomer at conditions effective to decyclize and polymerize said cyclic aryl-containing siloxane polymer and form a polysiloxane having a repeating unit of

29



30 wherein each  $R^4$  is independently selected from the group consisting of alkyl radicals, substituted alkyl radicals, aryl radicals and substituted aryl radicals.

35 20. The method of claim 19 wherein each  $R^4$  has 1 to about 3 carbon atoms and each  $R^2$  has 6 to about 10 carbon atoms.

21. The method of claim 19 wherein each  $R^4$  is methyl, each  $R^1$  has 1 to about 6 carbon atoms, and each  $R^2$  is phenyl.

40 22. The method of claim 19 wherein at least one end blocking group precursor is included in step (b) in an amount effective to provide said polysiloxane with end blocking groups.

5 23. The method of claim 19 wherein said cyclic hydride-containing siloxane monomer has a plurality of hydride groups, and step (a) is effective to chemically react said component with all of said hydride groups of said cyclic hydride-containing siloxane monomer.

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 93/03497

**I. CLASSIFICATION OF SUBJECT MATTER** (if several classification symbols apply, indicate all)<sup>6</sup>According to International Patent Classification (IPC) or to both National Classification and IPC  
Int.Cl. 5 C08G77/04; G02B1/04**II. FIELDS SEARCHED**Minimum Documentation Searched<sup>7</sup>

Classification System

Classification Symbols

Int.Cl. 5

G02B ; C08G

Documentation Searched other than Minimum Documentation  
to the Extent that such Documents are Included in the Fields Searched<sup>8</sup>**III. DOCUMENTS CONSIDERED TO BE RELEVANT<sup>9</sup>**

Category <sup>10</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>
X	FR,A,1 235 722 (DOW CORNING CORPORATION) 30 May 1960 see claim 1 see page 3, column 2, paragraph 3 see page 4; examples 2,3 see page 5; example 10 ---	1-5
A	US,A,4 618 665 (JOSEPH T. BRAUN) 21 October 1986 see claim 1 ---	1
A	DE,A,2 446 818 (DOW CORNING CORPORATION) 3 April 1975 see claim 1 see page 4, paragraph 3 - page 5, paragraph 2 ---	1
	--- -/-	

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- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step
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**IV. CERTIFICATION**

Date of the Actual Completion of the International Search

23 AUGUST 1993

Date of Mailing of this International Search Report

07.09.93

International Searching Authority

EUROPEAN PATENT OFFICE

Signature of Authorized Officer

DEPIJPER R.D.C.



III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category <sup>o</sup>	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No.
A	EP,A,0 202 542 (SUMITOMO ELECTRIC IND. LTD.) 26 November 1986 see claims 1,3 see page 6, line 10 - page 7, line 6 see page 10 - page 11; example 1 -----	1
A	JOURNAL OF POLYMER SCIENCE PART A1 vol. 9, no. 2, February 1971, pages 571 - 574 H.G. GOLLMAR 'preparation of some unsaturated silanes' -----	1

**ANNEX TO THE INTERNATIONAL SEARCH REPORT  
ON INTERNATIONAL PATENT APPLICATION NO.**

US 9303497  
SA 73312

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.  
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23/08/93

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
FR-A-1235722		None	
US-A-4618665	21-10-86	None	
DE-A-2446818	03-04-75	US-A- 3933678 AT-B- 342302 AU-A- 7344974 BE-A- 820587 CA-A- 1035567 FR-A, B 2272144 FR-A, B 2284640 GB-A- 1418453 JP-A- 50064354 JP-B- 51029906 NL-A- 7412702 SE-B- 420318 SE-A- 7412299 SE-A- 7804131	20-01-76 28-03-78 25-03-76 01-04-75 01-08-78 19-12-75 09-04-76 17-12-75 31-05-75 28-08-76 03-04-75 28-09-81 02-04-75 12-04-78
EP-A-0202542	26-11-86	JP-A- 61254624 JP-A- 61275330	12-11-86 05-12-86

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For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

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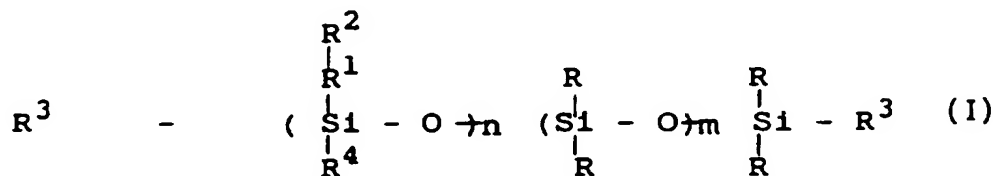
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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification <sup>5</sup> : <b>C08G 77/04, G02B 1/04</b></p>	<p><b>A1</b></p>	<p>(11) International Publication Number: <b>WO 93/21258</b> (43) International Publication Date: 28 October 1993 (28.10.93)</p>
<p>(21) International Application Number: PCT/US93/03497 (22) International Filing Date: 13 April 1993 (13.04.93) (30) Priority data: 07/868,412 14 April 1992 (14.04.92) US (71) Applicant: ALLERGAN, INC. [US/US]; 2525 Dupont Drive, Post Office Box 19534, Irvine, CA 92713-9534 (US). (72) Inventor: YANG, Shih-Liang, Stanley ; 25951 La Cuesta Avenue, Laguna Hills, CA 92653 (US). (74) Agents: VOET, Martin, A. et al.; Allergan, Inc., 2525 Dupont Drive, Post Office Box 19534, Irvine, CA 92713-9534 (US).</p>		<p>(81) Designated States: AU, CA, JP, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report.</i> <i>With amended claims.</i>  <b>Date of publication of the amended claims:</b> 25 November 1993 (25.11.93)</p>

(54) Title: POLYSILOXANES, METHODS OF MAKING SAME AND HIGH REFRACTIVE INDEX SILICONES MADE FROM SAME



(57) Abstract

A new class of polysiloxanes is disclosed. Such polysiloxanes have formula (I) wherein each R and R<sup>4</sup> is independently selected from alkyl radicals, substituted alkyl radicals, aryl radicals and substituted aryl radicals; each R<sup>1</sup> is independently selected from divalent radicals; each R<sup>2</sup> is independently selected from aryl radicals and substituted aryl radicals; each R<sup>3</sup> is independently selected from monovalent hydrocarbon radicals having a multiple bond and monovalent substituted hydrocarbon radicals having a multiple bond; n is integer in the range of about 6 to about 500; and m is an integer in the range of 0 to about 500. Methods for making such polysiloxanes are also disclosed. In addition, high refractive index, that is having a refractive index of at least about 1.46, silicone compositions useful in the fabrication of lens bodies are disclosed.

**FOR THE PURPOSES OF INFORMATION ONLY**

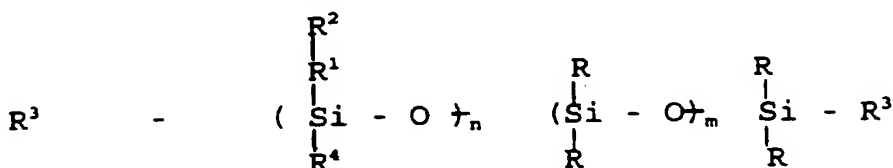
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FI	Finland				

## AMENDED CLAIMS

[received by the International Bureau on 26 October 1993 (26.10.93);  
original claims 1, 2, 5, 6 and 9 amended; new claims 24-38 added;  
remaining claims unchanged (9 pages)]

1. A polysiloxane having the formula:



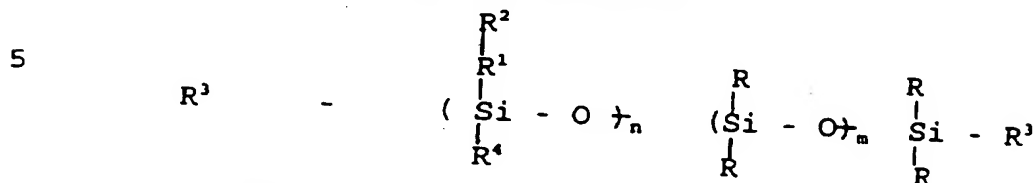
5 wherein each R and R<sup>4</sup> is independently selected from the group consisting of alkyl radicals, substituted alkyl radicals, aryl radicals and substituted aryl radicals; each R<sub>1</sub> is independently selected from the group  
10 consisting of divalent radicals; each R<sup>2</sup> is independently selected from aryl radicals and substituted aryl radicals; each R<sup>3</sup> is independently selected from the group consisting of monovalent hydrocarbon radicals having a multiple bond and substituted monovalent hydrocarbon  
15 radicals having a multiple bond; n is an integer in the range of about 6 to about 500; and m is an integer in the range of 0 to about 500, said polysiloxane has a refractive index of at least about 1.46, and the aryl-containing substituents in said polysiloxane comprise at  
20 least about 10 mol% of the total silicon-bound substituents in said polysiloxane.

2. The polysiloxane of claim 1 wherein each R<sup>4</sup> is selected from the group consisting of alkyl radicals and substituted alkyl radicals; each R<sup>2</sup> has 6 to about 10 carbon atoms; each R<sup>3</sup> has 2 to about 5 carbon atoms; and  
5 the aryl-containing substituents in said polysiloxane comprise at least about 15 mol% of the total silicon-bound substituents in said polysiloxane.

3. The polysiloxane of claim 1 wherein each R and R<sup>4</sup> is methyl, each R<sup>1</sup> has 1 to about 6 carbon atoms, each R<sup>2</sup> is phenyl and each R<sup>3</sup> is vinyl.

4. The polysiloxane of claim 3 wherein each  $R^1$  is a methylene radical or an ethylene radical.

5. A composition comprising:  
a cross-linked copolymer of (1) at least one polysiloxane having the formula



and (2) at least one cross-linker component, wherein each  $R$  and  $R^4$  is independently selected from the group consisting of alkyl radicals, substituted alkyl radicals, aryl radicals and substituted aryl radicals; each  $R^1$  is independently selected from the group consisting of divalent hydrocarbon radicals and divalent substituted hydrocarbon radicals; each  $R^2$  is independently selected from the group consisting of aryl radicals and substituted aryl radicals; each  $R^3$  is independently selected from the group consisting of monovalent hydrocarbon radicals having a multiple bond and substituted hydrocarbon radicals having a multiple bond;  $n$  is an integer in the range of about 6 to about 500; and  $m$  is an integer in the range of 0 to about 500, said composition being elastomeric and having a refractive index of at least about 1.46, and the aryl-containing substituents in said polysiloxane comprise at least about 10 mol% of the total silicon-bound substituents in said polysiloxane.

6. The composition of claim 5 wherein the aryl-containing substituents in said polysiloxane comprise at least about 20 mol% of the total silicon-bound substituents in said polysiloxane.

7. The composition of claim 5 which further comprises at least one reinforcer component in said copolymer in an amount effective to increase the strength of said composition.

8. The composition of claim 5 wherein said cross-linker component is a poly organo hydrosiloxane.

9. The composition of claim 5 wherein each R has 1 to about 4 carbon atoms; each R<sup>2</sup> has 6 to about 10 carbon atoms; each R<sup>3</sup> has 2 to about 5 carbon atoms; and each R<sup>4</sup> is independently selected from the group consisting of  
5 alkyl radicals and substituted alkyl radicals.

10. The composition of claim 5 wherein each R and R<sup>4</sup> is methyl, each R<sup>1</sup> has 1 to about 6 carbon atoms, each R<sup>2</sup> is phenyl and each R<sup>3</sup> is vinyl.

11. The composition of claim 8 wherein said poly organo hydrosiloxane is selected from the group consisting of copolymers of methylhydrosiloxanes and phenylmethylsiloxane, copolymers of methylhydrosiloxane  
5 and diphenylsiloxane, and mixtures thereof.

12. A lens body for use in or on a mammalian eye comprising the composition of claim 5.

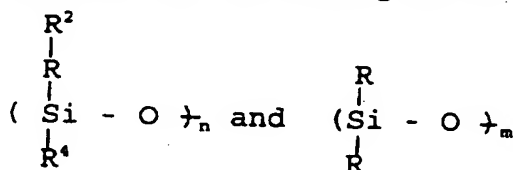
13. A lens body for use in or on a mammalian eye comprising the composition of claim 7.

14. A method for making a polysiloxane comprising:  
(a) contacting a cyclic hydride-containing siloxane monomer with at least one component having the formula

5  $R^2 - f$

wherein  $R^2$  is selected from the group consisting of aryl radicals and substituted aryl radicals, and F is a functional monovalent radical capable of reacting with a hydride group of said cyclic hydride-containing siloxane monomer at conditions effective to chemically react said component with at least one of said hydride groups of said cyclic hydride-containing siloxane monomer and form a cyclic aryl-containing siloxane monomer including at least one -  $R^1$  -  $R^2$  group wherein  $R^1$  is selected from the group consisting of divalent radicals; and

(b) contacting said cyclic aryl-containing siloxane monomer with at least one siloxane monomer at conditions effective to decyclize and polymerize said cyclic aryl-containing siloxane monomer and form a polysiloxane having the following units



wherein each R and  $R^4$  is independently selected from the group consisting of alkyl radicals, substituted alkyl radicals, aryl radicals and substituted alkyl radicals, n is an integer in the range of 6 to about 500 and m is an integer in the range of 1 to about 500.

15. The composition of claim 14 wherein each R has 1 to about 3 carbon atoms; each  $R^2$  has 6 to about 10 carbon atoms; each  $R^4$  is independently selected from the group consisting of alkyl radicals and substituted alkyl radicals.

16. The method of claim 14 wherein each R and  $R^4$  is methyl, each  $R^1$  has 1 to about 6 carbon atoms, and each  $R^2$  is phenyl.



17. The method of claim 14 wherein at least one end blocking group precursor is included in step (b) in an amount effective to provide said polysiloxane with end blocking groups.

18. The method of claim 14 wherein said cyclic hydride-containing siloxane monomer has a plurality of hydride groups, and step (a) is effective to chemically react said component with all of said hydride groups of  
5 said cyclic hydride-containing siloxane monomer.

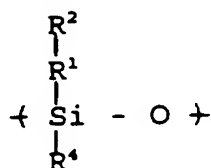
19. A method for making a polysiloxane comprising:  
(a) contacting a cyclic hydride-containing siloxane monomer with at least one component having the formula

5



wherein  $R^2$  is selected from the group consisting of aryl radicals and substituted aryl radicals, and  $f$  is a functional monovalent radical capable of reacting with a hydride group of said cyclic hydride-containing siloxane monomer at conditions effective to chemically react said  
10 component with at least one of said hydride groups of said cyclic hydride-containing siloxane monomer and form a cyclic aryl-containing siloxane monomer including at least one -  $R^1 - R^2$  group wherein  $R^1$  is selected from the group  
15 consisting of divalent radicals; and

(b) contacting said cyclic aryl-containing siloxane monomer at conditions to decyclize and polymerize said cyclic aryl-containing siloxane monomer, react said siloxane polymer and form a polysiloxane having a  
20 repeating unit of



25 wherein each R<sup>4</sup> is independently selected from the group consisting of alkyl radicals, substituted alkyl radicals, aryl radicals and substituted alkyl radicals.

20. The method of claim 19 wherein each R<sup>4</sup> has 1 to about 3 carbon atoms and each R<sup>2</sup> has 6 to about 10 carbon atoms.

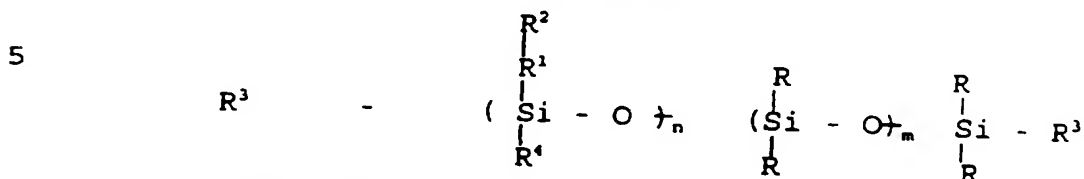
21. The method of claim 19 wherein each R<sup>4</sup> is methyl, each R<sup>1</sup> has 1 to about 6 carbon atoms, and each R<sup>2</sup> is phenyl.

22. The method of claim 19 wherein at least one end blocking group precursor is included in step (b) in an amount effective to provide said polysiloxane with end blocking groups.

23. The method of claim 19 wherein said cyclic hydride-containing siloxane monomer has a plurality of hydride groups, and step (a) is effective to chemically react said component with all of said hydride groups of  
5 said cyclic hydride-containing siloxane monomer.

24. The composition of claim 5 which is elastomeric.

25. A composition comprising:  
a cross-linked copolymer of (1) at least one polysiloxane having the formula



and (2) at least one cross-linker component, wherein each R and R<sup>4</sup> is independently selected from the  
10 group consisting of alkyl radicals, substituted alkyl

radicals, aryl radicals and substituted aryl radicals; each  $R^1$  is independently selected from the group consisting of divalent hydrocarbon radicals and divalent substituted hydrocarbon radicals; each  $R^2$  is independently  
15 selected from the group consisting of aryl radicals and substituted aryl radicals; each  $R^3$  is independently selected from the group consisting of monovalent hydrocarbon radicals having a multiple bond and substituted hydrocarbon radicals having a multiple bond;  
20  $n$  is an integer in the range of about 6 to about 500; and  $m$  is an integer in the range of 0 to about 500, said composition being elastomeric and having a refractive index of at least about 1.46, and the aryl-containing substituents in said polysiloxane comprise at least about  
25 10 mol% of the total silicon-bound substituents in said polysiloxane.

26. The composition of claim 25 wherein the aryl-containing substituents in said polysiloxane comprise at least about 15 mol% of the total silicon-bound substituents in said polysiloxane.

27. The composition of claim 25 which further comprises at least one reinforcer component in said copolymer in an amount effective to increase the strength of said composition.

28. The composition of claim 25 wherein said cross-linker component is a poly organo hydrosiloxane.

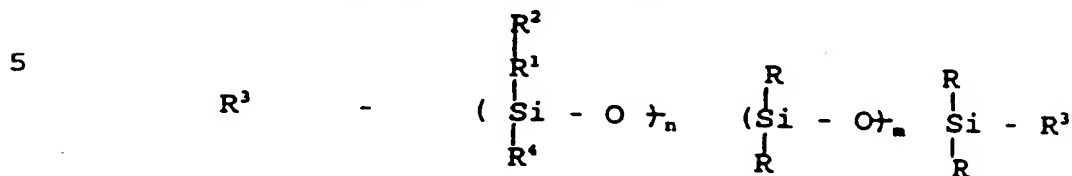
29. The composition of claim 25 wherein each  $R$  has 1 to about 4 carbon atoms; each  $R^2$  has 6 to about 10 carbon atoms; each  $R^3$  has 2 to about 5 carbon atoms; and each  $R^4$  is independently selected from the group  
5 consisting of alkyl radicals and substituted alkyl

radicals.

30. The method of claim 23 wherein each R and R<sup>4</sup> is methyl, each R<sup>1</sup> has 1 to about 6 carbon atoms, and each R<sup>2</sup> is phenyl and each R<sup>3</sup> is vinyl.

31. The composition of claim 25 wherein the aryl-containing substituents in said polysiloxane comprise at least about 20 mol% of the total silicon-bound substituents in said polysiloxane.

32. A composition comprising:  
a cross-linked copolymer of (1) at least one polysiloxane having the formula



and (2) at least one cross-linker component, wherein each R and R<sup>4</sup> is independently selected from the group consisting of alkyl radicals, substituted alkyl radicals, aryl radicals and substituted aryl radicals; each R<sup>1</sup> is independently selected from the group consisting of divalent hydrocarbon radicals and divalent substituted hydrocarbon radicals; each R<sup>2</sup> is independently selected from the group consisting of aryl radicals and substituted aryl radicals; each R<sup>3</sup> is independently selected from the group consisting of monovalent hydrocarbon radicals having a multiple bond and substituted hydrocarbon radicals having a multiple bond; n is an integer in the range of about 6 to about 500; and m is an integer in the range of 0 to about 500, said composition being elastomeric and having a refractive index of at least about 1.46, and the aryl-containing substituents in said polysiloxane comprise at least about

25 20 mol% of the total silicon-bound substituents in said polysiloxane.

33. The composition of claim 32 which further comprises at least one reinforcer component in said copolymer in an amount effective to increase the strength of said composition.

34. The composition of claim 32 wherein said cross-linker component is a poly organo hydrosiloxane.

35. The composition of claim 32 wherein each R has 1 to about 4 carbon atoms; each R<sup>2</sup> has 6 to about 10 carbon atoms; each R<sup>3</sup> has 2 to about 5 carbon atoms; and each R<sup>4</sup> is independently selected from the group  
5 consisting of alkyl radicals and substituted alkyl radicals.

36. The composition of claim 32 wherein each R and R<sup>4</sup> is methyl, each R<sup>1</sup> has 1 to about 6 carbon atoms, each R<sup>2</sup> is phenyl and each R<sup>3</sup> is vinyl.

37. The composition of claim 37 which is elastomeric.

38. The composition of claim 32 which has a refractive index of at least about 1.50.

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